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(54) Abstract Title
Polychromatic,organic electroluminescent devices

(57) The polychromatic EL device includes a transparent substrate 7; a colour conversion filter including fluorescent layers 4, 5 and 6 disposed in a planar arrangement on the substrate 7 and spaced apart from each other; and an organic EL element above the colour conversion filter including an organic light-emitting layer 11. A film laminate is interposed between the colour conversion filter and the organic EL element. The film laminate include a transparent base film 1; a hard coat layer 3 on the upper surface of the transparent base film 1 and a coupling layer 2 on the lower surface of the transparent base film 1. The hard coat layer 3 exhibits a gas blocking capability and hardness enough to protect the filter during the formation of the organic EL element. The coupling layer 2 is made of a photo-curing resin or a thermosetting resin which hardens at 150°C or lower.

Fig. 3

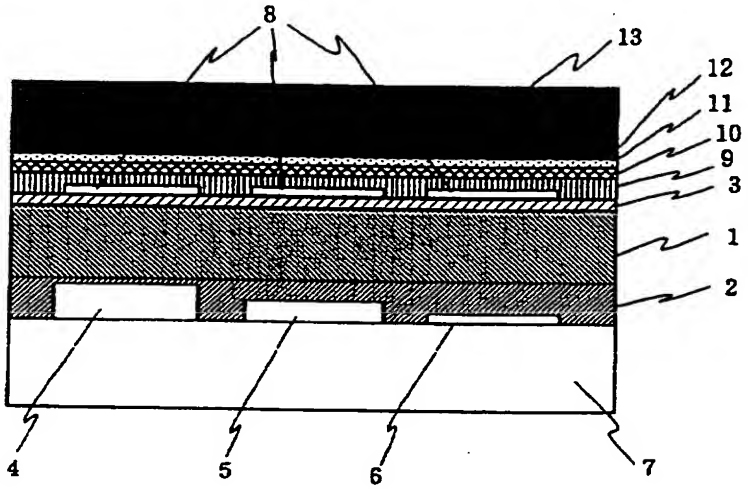

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Fig. 1

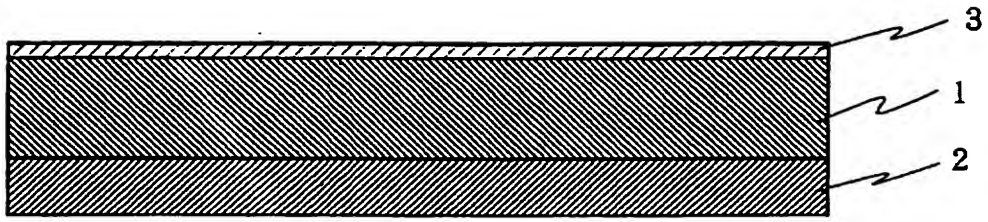


Fig. 2

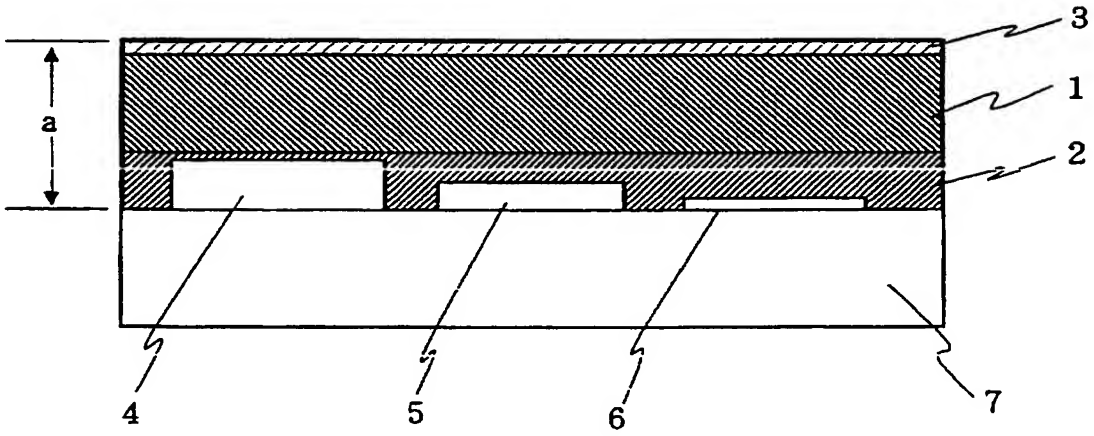


Fig. 3

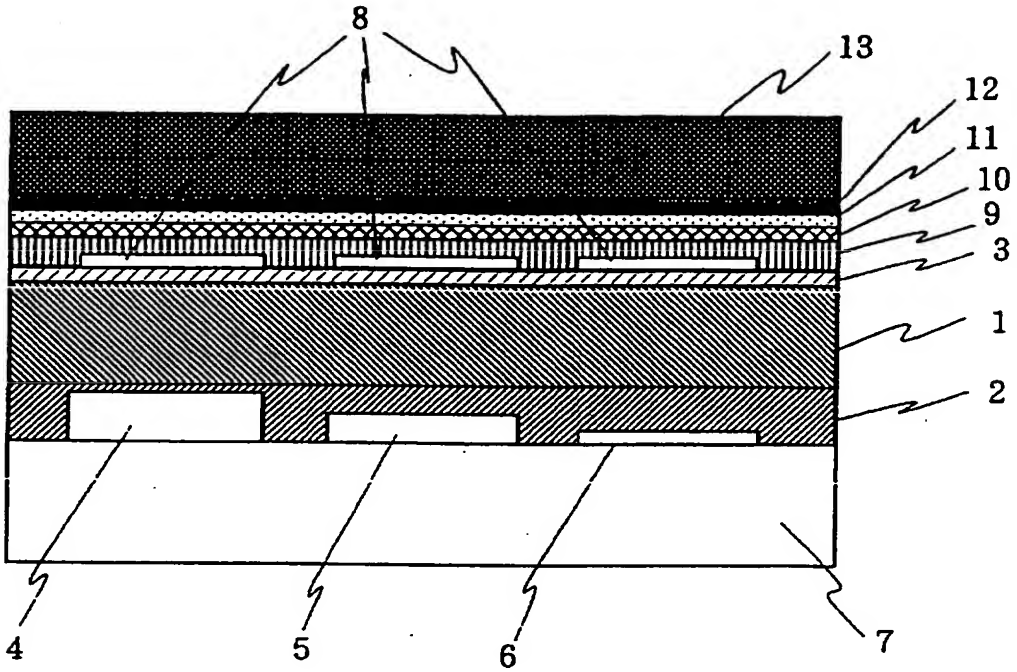


Fig. 4

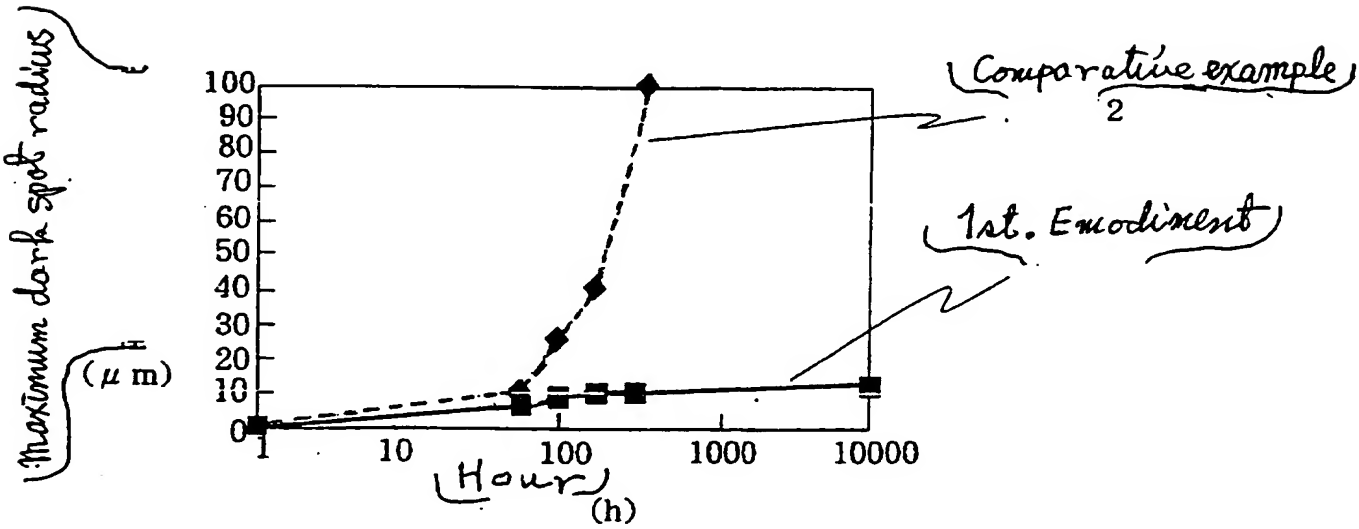


Fig. 5

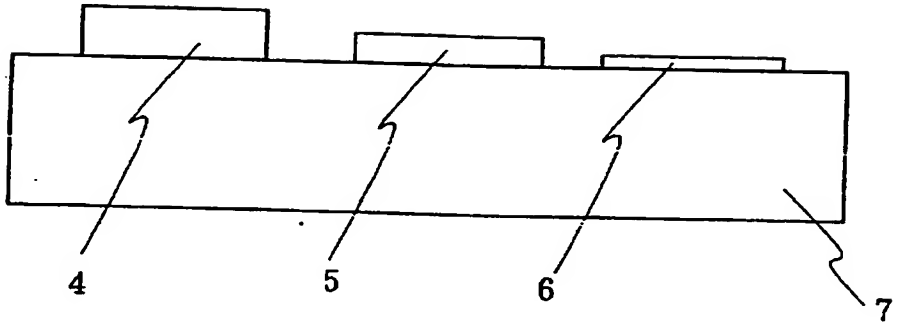


Fig. 6

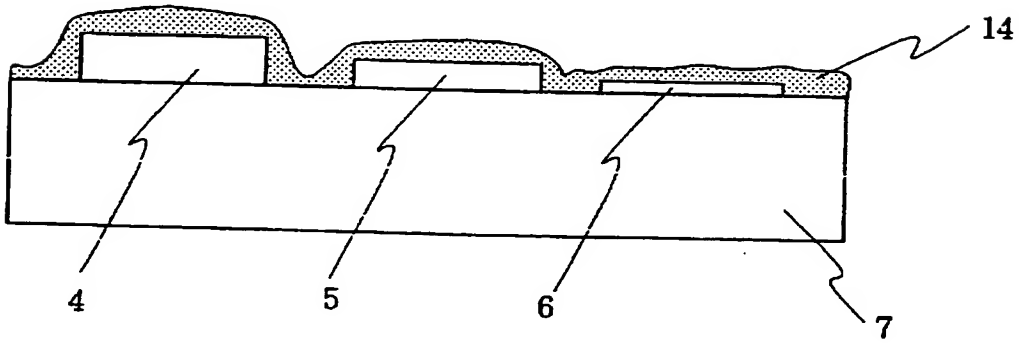


Fig. 7

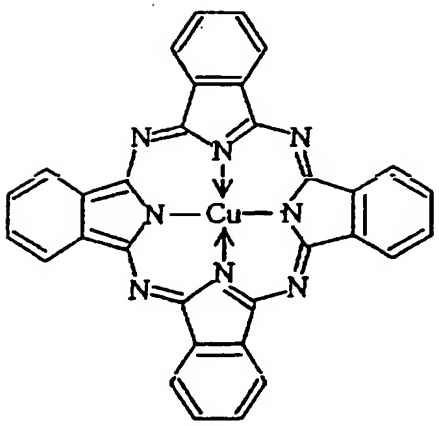


Fig. 8

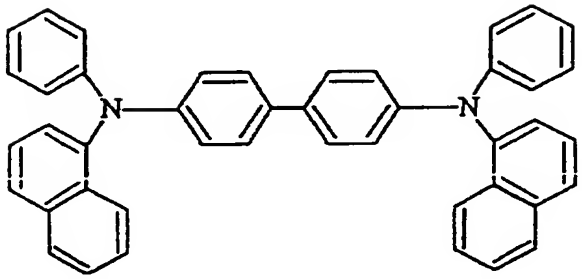


Fig. 9

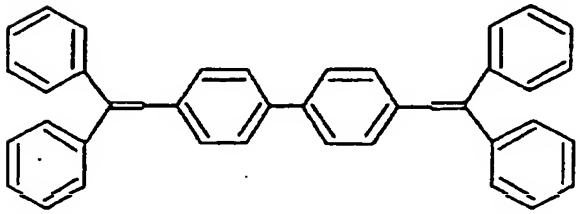
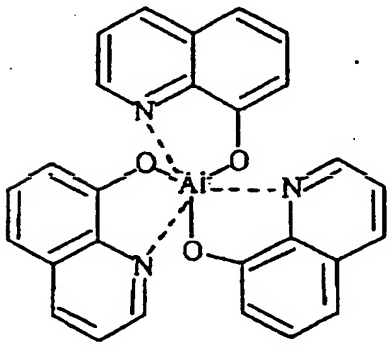


Fig. 10



A POLYCHROMATIC ORGANIC ELECTROLUMINESCENT DEVICE AND A METHOD OF MANUFACTURING THE SAME

The present invention relates to a polychromatic organic electroluminescent (hereinafter referred to as "EL") device used in the organic EL display and such display devices. The present invention relates also to a method of manufacturing the polychromatic organic EL device.

Since Tang et al. reported a laminate-type EL device which facilitated obtaining high luminance of 1000 cd/m^2 at a low applied voltage of 10 V (cf. Appl. Phys. Lett., 51, 913 (1987)), developments of organic EL devices have been conducted vigorously. The organic EL device, which is a thin-film device of self-light-emitting type, has been expected to be applicable to flat-panel displays by virtue of its characteristic features such as a low drive voltage, high resolution and wide field of view. From the view point of wider application, it is required for the organic EL device to emit polychromatic light.

Conceivable device structures for realizing polychromatic display include planar arrangement of EL elements, each emitting light in one of the three primary colours (a red-light-emitting EL element, a green-light-emitting EL element and a blue-light-emitting EL element), in a predetermined pattern, and arrangement of three colour filters, each for each of the three primary colours (a colour filter for making red light pass, a colour filter for making green light pass and a colour filter for making blue light pass), on a white-light-emitting element.

However, the process of patterning the organic EL elements lowers the light-emitting efficiency, complicates the manufacturing process so much and makes mass production of the device very difficult. And, any light-emitting element which emits stable white light bright enough to be employed for the colour filter method has not been obtained yet.

Recently, a colour conversion method which uses filters, each made of a fluorescent material which absorbs the light from the organic EL element and emits certain fluorescent light in the visible region, has been proposed (cf. Japanese Unexamined Laid Open Patent Applications No. H03-152897 and No. H05-258860). Since the light from the organic EL element compatible with the colour conversion method is not limited to white one, an organic light-emitting element which emits brighter light may be used as a light source. In the colour conversion method which uses an organic EL element which emits blue light, the conversion efficiency to the longer wavelength exceeds 60%.

As far as the present inventors have examined, the fluorescent materials used for converting the light from the organic EL element to the light of a desired wavelength are so sensitive to the light of a specific wavelength, moisture, heat and organic solvents that the fluorescent materials are decomposed easily by these causes and lose the required functions. Therefore, the polychromatic light-emitting device based on the colour conversion method subjects to various limitations in configuration and manufacture thereof.

In detail, the Japanese Unexamined Laid Open Patent Application No. H08-279394 (Hereinafter referred to as the "above identified patent application") has proposed a polychromatic light-emitting device which protects the fluorescent materials with an insulative inorganic oxide layer. According to this conventional device structure, the fluorescent layers are protected by the insulative inorganic oxide layer interposed between the fluorescent layers and an organic EL element. The above identified patent application also discloses that it is preferable to interpose a fluorescent protection layer and an adhesive layer between fluorescent layers and the insulative inorganic oxide layer. The above identified patent application discloses the following four laminate structures.

(1) A transparent substrate / fluorescent layers / a transparent and electrically insulative inorganic oxide layer / an organic EL element

(2) A transparent substrate / fluorescent layers / an adhesive layer / a transparent and electrically insulative inorganic oxide layer / an organic EL element

(3) A transparent substrate / fluorescent layers / a protection layer (transparent flattening layer) / an adhesive layer / a transparent and electrically insulative inorganic oxide layer / an organic EL element

(4) A transparent substrate / fluorescent layers / a protection layer (transparent flattening layer) / a transparent and electrically insulative inorganic oxide layer// an organic EL element

In a colour filter which uses fluorescent materials (hereinafter referred to as a "colour conversion filter"), the fluorescent layers are different in thickness for balancing the colours due to the different conversion efficiencies of the fluorescent materials for converting to the respective colours. As shown in Figure 5, the thickness differences cause steps in the surface of the colour conversion filter opposite to a glass substrate 7. According to the experimental results by the present inventors, the conversion efficiency of the converter material for converting to red (hereinafter simply referred to as the "red fluorescent material") is lower than the conversion efficiencies of the other converter materials (the green fluorescent material and the blue fluorescent material). If one wants to obtain emitted light with high colour purity, a red fluorescent layer 4 will be several tens μ m in thickness. As a result, the steps of 10 μ m or more will be caused between the red fluorescent layer 4 and a green fluorescent layer 5 of from 4 to 10 μ m in thickness as well as between the red fluorescent layer 4 and a blue fluorescent layer 6 of from 0 to 5 μ m in thickness. When the EL element emits blue light, the blue fluorescent layer 6 is not disposed, that is the thickness of the blue fluorescent layer 6 is 0 μ m.

It is difficult for the device structure disclosed in the Japanese Unexamined Laid Open Patent Application No. H08-279394 which forms the transparent and insulative inorganic oxide layer directly on the fluorescent layers to flatten the steps of the colour conversion

filter. Therefore, concave and convex portions are inevitably caused also on the insulative inorganic oxide layer 14 as shown in Figure 6. The concave and convex portion, thus caused, adversely affects the colour purity and the angle of visibility.

In forming the protection layer and the adhesive layer one by one between the fluorescent layers and the transparent and insulative inorganic oxide layer, the fluorescent materials are exposed repeatedly to heat and exposure light in curing the protection layer and the adhesive layer. It is difficult to completely prevent the fluorescent materials from deterioration caused by the repeated exposure to the heat and the light. Resin coating on a wafer having high steps such as the colour conversion filter causes thin portions in the resin coat. For maintaining the protective function while avoiding formation of these thin portions, it is necessary to thicken the protection layer. With increasing thickness of the protection layer, the colour conversion filter and the EL element are spaced more apart. As the distance between the colour conversion filter and the EL element is elongated, the angle of visibility is narrowed. Moreover, since the fluorescent materials are so sensitive to chemical reagents, moisture, light and heat that the materials for the protection layer and the method for forming the protection layer are limited. Since very complicated manufacturing steps are necessary to form the above described layer structures, the conventional device structures are industrially disadvantageous.

In view of the foregoing, it is an object of the invention to provide a polychromatic EL device which exhibits a wide angle of visibility. It is another object of the invention to provide apolychromatic EL device which prevents the fluorescent materials from deterioration in the manufacturing process thereof, It is still another object of the invention to provide a method of manufacturing such a polychromatic EL device.

According to an aspect of the invention, there is provided a polychromatic organic electroluminescent device which includes a colour conversion filter including a transparent substrate and fluorescent layers formed on the transparent substrate and spaced apart from each other; an organic electroluminescent element including an organic light-emitting layer; and a functional film laminate interposed between the colour conversion filter and the organic electroluminescent element; the colour conversion filter and the organic electroluminescent element being arranged such that the fluorescent layers absorb the light from the organic light-emitting layer and emit light in their respective colours; the functional film laminate including a transparent base film, a transparent hard coat layer on the upper surface of the transparent base film and a coupling layer on the lower surface of the transparent base film; the hard coat layer being transparent for the visible light, the hard coat layer exhibiting a gas blocking capability and hardness enough to endure the formation of the organic electroluminescent element on its upper surface; the coupling layer being made of a photo-curing resin or a thermosetting resin which hardens at 150°C or lower.

Advantageously, one of the fluorescent layers is replaced by a colour filter for adjusting the colour and the colour purity of the light emitted from the organic light-emitting layer.

Advantageously, the functional film laminate further includes transparent electrodes on the hard coat layer.

Advantageously, the hard coat layer is a resin layer or an inorganic oxide layer, the resin layer and the inorganic oxide layer exhibiting pencil hardness H or harder.

Advantageously, the coupling layer contains a resin which hardens by the visible light.

Advantageously, the coupling layer contains a resin which thermally cross-links at 150°C or lower.

Advantageously, the base film is from 1 to 50 μ m in thickness.

Advantageously, the fluorescent layers include a red fluorescent layer which converts the light from the organic electroluminescent element to red light, a green fluorescent layer which converts the light from the organic electroluminescent element to green light and a blue fluorescent layer which converts the light from the organic electroluminescent element to blue light.

According to another aspect of the invention, there is provided a method of manufacturing a polychromatic organic electroluminescent which includes a colour conversion filter including a transparent substrate and fluorescent layers formed on the transparent substrate and spaced apart from each other; an organic electroluminescent element including an organic light-emitting layer; and a functional film laminate interposed between the colour conversion filter and the organic electroluminescent element; the colour conversion filter and the organic electroluminescent element being arranged such that the fluorescent layers absorb the light from the organic light-emitting layer and emit light in their respective colours; the functional film laminate including a transparent base film, a transparent hard coat layer on the upper surface of the transparent base film and a coupling layer on the lower surface of the transparent base film; the hard coat layer being transparent for the visible light, the hard coat layer exhibiting a gas blocking capability and hardness enough to endure the formation of the organic electroluminescent element on its upper surface; the coupling layer being made of photo-curing resin or a thermosetting resin which hardens at 150°C or lower, which methodincludes the step of bonding the functional film laminate onto the upper surface of the colour conversion filter; and the step of arranging the organic electroluminescent element on the upper surface of the functional film laminate.

The fluorescent materials are prevented from deterioration according to the invention, since the colour conversion filter is not repeatedly exposed to heat or light like in curing the protection and adhesive layers of the prior art. Therefore, the colour conversion filter keeps exhibiting an excellent colour conversion capability. The differences between the levels of the fluorescent layers (a red fluorescent layer, a green fluorescent layer and a blue fluorescent layer), spaced apart from each other on the planar substrate, are smoothed out by the thinnest coupling layer in the functional film laminate. By disposing the organic EL element directly onto the functional film laminate, a colour display device in which the spacing between the colour conversion filter and the organic EL element is short, that is a colour display device which exhibits a wide angle of visibility, is obtained. And, according to the manufacturing method of the invention, the colour conversion filter is protected simply by bending the functional film laminate onto the colour conversion filter.

Now the present invention will be explained hereinafter with reference to the accompanied drawing Figures, in which:

Figure 1 is a cross-section of a functional film laminate according to the invention for protecting a colour conversion filter including fluorescent layers arranged on a transparent substrate according to the invention;

Figure 2 is a cross-section of the colour conversion filter, the surface thereof is protected by the functional film laminate according to the invention;

Figure 3 is a cross-section of the polychromatic organic EL device according to the invention;

Figure 4 is a pair of curves plotting the maximum dark spot sizes in the devices according to one of the embodiments and one of the comparative examples with elapse of time;

Figure 5 is a cross-section of a conventional colour conversion filter;

Figure 6 is another cross-section of the conventional colour conversion filter protected by an inorganic oxide layer;

Figure 7 describes the structural formula of copper phthalocyanine (CuPc), used for the hole injection layer according to the invention;

Figure 8 describes the structural formula of 4,4'-bis[N-(1-naphthyl)-N- phenylamino]biphenyl (α -NPD) used for the hole transport layer according to the invention;

Figure 9 4,4'-bis(2,2-diphenylvinyl)biphenyl (DPVBi) used for the light-emitting layer according to the invention; and

Figure 10 describes the structural formula of aluminium chelate (Alq) used for the electron injection layer according to the invention.

Figure 1 is a cross-section of a functional film laminate according to the invention for protecting a colour conversion filter including fluorescent layers arranged on a transparent substrate according to the invention. Figure 2 is a cross-section of the colour conversion filter, the surface thereof is protected by the functional film laminate according to the invention.

Referring now to Figure 1, the functional film laminate according to the invention includes a transparent base film 1, a hard coat layer 3 on the upper surface of the transparent base film 1 and a coupling layer 2 on the lower surface of the transparent base film 1. The hard coat layer 3 well blocks gases used in forming an EL element thereon. The hard coat layer 3 is so hard that the EL element may be formed thereon with no problems. The hard coat layer 3 is transparent for the visible light. The coupling layer is made of a photo-curing resin or a thermosetting resin which thermally hardens at 150°C or lower. The functional film laminate is bonded onto a colour conversion

filter as shown in Figure 2 to protect fluorescent layers 4, 5 and 6 (a red fluorescent layer 4, a green fluorescent layer 5 and a blue fluorescent layer 6) separated from each other on a flat transparent substrate 7. Either one of the fluorescent layers 4, 5 and 6 may be replaced by a colour filter for adjusting the colour and the colour purity of the light emitted from an organic light-emitting layer.

Preferably, the base film 1 is made of a stable resin that does not yield any chemical species which may deteriorate the fluorescent materials and the organic EL element between the room temperature and 150°C. Preferably, the base film 1 is from 1 to 50 μm in thickness and, more preferably, from 5 to 20 μm in thickness. When the base film 1 is thinner than 1 μm, it is difficult to flatten the colour conversion filter, the functional film laminate is cut easily and the workability becomes worse. Although it depends on the refractive index of the material, the light scattered from a fluorescent layer leaks to the adjacent fluorescent layers which are not emitting light and the angle of visibility is narrowed, when the base film 1 is thicker than 50 μm.

The materials for the base film 1 have no specific limitations as far as they are formed to be a film easily, well transparent, for example exhibiting the transparency of 50% or more in the wavelength range between 400 nm and 700 nm, and resistant against the heatgenerated in the next manufacturing steps and in driving the device. For example, poly(ethylene terephthalate) (PET), polyethylene (PE), polypropylene (PP), polycarbonate (PC), polyacrylate (PA), poly(ether sulfone) (PES), poly(vinyl fluoride) (PFV) and polyetheretherketone (PEEK) are used for the base film 1.

The coupling layer 2 of the functional film laminate is made of a polymer resin and such a transparent material which do not deteriorate the fluorescent materials. The coupling layer 2 is disposed primarily for flattening the colour conversion filter without adversely affecting the colour conversion characteristics. The coupling layer 2 is disposed also as a buffer between the colour conversion filter and the base film 1.

Preferably, the coupling layer 2 exhibits high adhesiveness and high transparency, for example the transparency of 50% or more in the wavelength range between 400 nm and 700 nm, and is formed as a coating film of μ m order in thickness on the base film 1. Any material, which does not dissolve the materials of the colour conversion filter, such as crosslinking-type urethane resins, olefin resins, acrylic resins, epoxy resins, melamine resins, phenolic resins, alkyd resins, ester resins and amide resins may be used for the coupling layer 2. The method for curing the coupling layer 2 has no limitations. Thermosetting, moisture-curing, chemical curing and photo-curing may be used alone or in an appropriate combination. The preferable temperature for the thermosetting is up to around 70°C for avoiding deterioration of the fluorescent materials. Preferably, the photo-curing is conducted with the visible light considering the deterioration of the fluorescent materials. The coupling layer may be coated on the base film by any conventional coating method such as the conventional spin-coating method, roll-coating method and cast-coating method.

The hard coat layer 3 of the functional film laminate is disposed on the side of the organic EL element of the base film 1. The hard coat layer 3 works as a barrier against gases and organic solvents. The hard coat layer 3 is disposed for protecting the materials of the colour conversion filter from the low molecular compounds such as moisture and resin monomers in the process of forming the organic EL element, including anodes, organic layers and cathodes, and for protecting the organic EL element formed on the functional film laminate by blocking moisture, oxygen, monomers and such low molecular compounds yielded from the colour conversion filter by deterioration with age or by heat generated in driving the device. The hard coat layer 3 is disposed also to facilitate forming the anodes of the organic EL element on the functional film laminate.

Preferably, the materials for the hard coat layer 3 exhibits a capability of blocking gases and organic solvents and high transparency. Preferably, the materials for the hard coat layer 3 facilitate forming hard coat layer 3 of from nm to μ m order in

thickness on the base film 1. It is also preferable for the hard coat layer 3 to exhibit the hardness H, more preferably the hardness 2H, in the pencil hardness test hard enough to endure the formation of the anodes. For example, polymers such as ester resins, epoxy resins, urethane resins and melamine resins, and inorganic oxides such as aluminium oxide, boric acid, barium oxide, germanium oxide, lithium oxide, magnesium oxide, lead oxide, silicon oxide, titanium oxide, zinc oxide and zirconia are used as the materials for the hard coat layer 3.

The colour conversion filter may be constructed in the conventional structure with the conventional materials by anyone of the conventional manufacturing methods. For example, the colour conversion filter includes a red fluorescent layer 4, a green fluorescent layer 5 and a blue fluorescent layer 6 spaced apart from one another and arranged on a planar substrate 7 such as a glass substrate. There exists no limitation in selecting the method for forming the colourconversion filter. For example, photolithography and micell electrolysis may be employed for forming the colour conversion filter.

The organic EL element to be arranged on the functional film laminate may be constructed in the conventional structure with the conventional materials by anyone of the conventional manufacturing methods. It is preferable for the organic EL element to emit light over the wavelength range between near ultraviolet and bluish green, though not always limited thereto.

Now the invention will be explained in connection with the preferred embodiments thereof.

A polychromatic organic EL device as schematically shown in Figure 3 was fabricated through the following steps.

A layer of blue material for a colour filter (Color Mosaic CB-7001 supplied from FUJIFILM OLIN CO., LTD.) was coated on a fusion-glass substrate 7 (143 mmX112 mmX 1.1mm) by spin-coating, and a line pattern of blue fluorescent layers 6, each 2 mm in length and 1 μm in thickness, spaced apart for 5.5 mm was obtained by

photolithographic patterning of the as coated layer. Then, layers of a mixture of Coumarin 6 (supplied from Aldrich Chemical Co., Inc.) and poly(vinyl chloride) were printed on the substrate 7 by screen printing, and a line pattern of green fluorescent layers 5, each 2 mm in length and 10 μ m in thickness, spaced apart for 5.5 mm was obtained by baking the as printed layers at 150°C. Then, layers of a mixture of Rhodamine 6G (supplied from Aldrich Chemical Co., Inc,) and poly(vinyl chloride) were printed on the substrate 7 by screen printing, and a line pattern of red fluorescent layers 4, each 2 mm in length and 30 μ m in thickness, spaced apart for 5.5 mm was obtained by baking the as printed layers at 100°C.

A hard coat layer 3 was deposited by vacuum deposition of a silicon compound on a surface of a base film 1 (High Barrier Film: MOS-TH, 12 μ m in thickness, supplied from Tokyo Oike Industry, Co., Ltd.). And, a coupling layer 2 was formed by roll-coating of a urethane resin (S720-Q supplied from Hodogaya Chemical Co., Ltd.) with the aid of an isocyanate crosslinking agent (A100 supplied from Nippon Polyurethane Industry Co., Ltd.) on another surface of the base film 1. Thus, a functional film laminate was formed.

The functional film laminate formed as described above was coated by roll-coating on the colour conversion filter and baked at 70°C for 2 hr. to harden the coupling layer 2.

An organic EL film on the colour conversion filter was formed in a hexa-layer structure including anodes 8, a hole injection layer 9, a hole transport layer 10, a light-emitting layer 11, an electron injection layer 12 and cathodes 13 as shown in Figure 3,

At first, an ITO layer for the anodes (transparent electrodes) 8 was deposited by sputtering on the entire upper surface of the functional film laminate bonded onto the colour conversion filter. A stripe pattern of the transparent electrodes 8, each 2 mm in length and 100 nm in thickness, spaced apart for 0.5 mm was obtained by photolithographic processing of a photoresist agent (OFPR-800 supplied from Tokyo Ohka Kogyo Co., Ltd.) coated on the ITO layer.

Then, the wafer was loaded in a vacuum deposition chamber of resistance heating type, and the hole injection layer 9, the hole transport layer 10, the light-emitting layer 11 and the electron injection layer 12 were formed one by one without breaking the vacuum of the chamber. In forming these layers, the pressure inside the chamber was reduced down to $1X10^{-4}$ Pa. The hole injection layer 9 was formed by depositing copper phthalocyanine (CuPc), described by the structural formula in Figure 7, to the thickness of 100 nm.

The hole transport layer 10 was formed by depositing 4,4′-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (α -NPD), described by the structural formula in Figure 8, to the thickness of 20 nm.

The light-emitting layer 11 was formed by depositing 4,4′-bis(2,2-diphenylvinyl)biphenyl (DPVBi), described by the structural formula in Figure 9, to the thickness of 30 nm.

The electron injection layer 12 was formed by depositing aluminium chelate (Alq), described by the structural formula in Figure 10, to the thickness of 20 nm.

Then, the wafer was taken out from the vacuum chamber. A mask for obtaining a stripe pattern of the cathodes 13, each 2 mm in length, spaced apart for 0.5 mm was attached onto the wafer so that the cathode stripes may extend perpendicularly to the ITO stripes. The wafer with the mask attached thereon was loaded again into the vacuum chamber and the cathodes 13 of Mg and Ag (10:1 in weight ratio) were deposited to the thickness of 200 nm.

The functional film laminate according to the second embodiment was fabricated in the similar manner as the functional film laminate of the first embodiment except that an epoxy acrylate resin doped with 1 weight % of a visible-light curing agent (IRGACURE-1850 (R.T.M.) supplied from Ciba Geigy AG.) was used for the coupling layer 2 in the second embodiment. The functional film laminate was bonded onto the fluorescent layers of the colour conversion filter. Then, the coupling layer was cured by irradiating 3000 mJ of the light from a

high-pressure mercury lamp, the short wavelength component of which (shorter than 400 nm) had been cut with a polycarbonate filter. Finally, an organic EL element, including anodes 8, a hole injection layer 9, a hole transport layer 10, a light-emitting layer 11, an electron injection layer 12 and cathodes 13, was formed on the uppersurface of the functional film laminate in the same manner as in the first embodiment.

A protection layer was formed on the surface of the colour conversion filter, on which the fluorescent layers had been formed, by coating a photo-curing adhesive of a methacrylate oligomer (3112 supplied from Three Bond Co., Ltd.), by exposing the coated adhesive to 3000 mJ of the 365 nm light and by baking the cured adhesive at 80°C.

Then, an organic EL element, including anodes 8, a hole injection layer 9, a hole transport layer 10, a light-emitting layer 11, an electron injection layer 12 and cathodes 18, was formed on the upper surface of the protection layer in the same manner as in the first embodiment.

The functional film laminate according to the comparative example 2 was formed in the similar manner as the functional film laminate of the first embodiment except that a PET film (12 μ m in thickness) on which any silicon compound had not been deposited was used for the base film 1. The functional film laminate was bonded onto the colour conversion filter in the same manner as in the first embodiment. Finally, an organic EL element, including anodes 8, a hole injection layer 9, a hole transport layer 10, a light-emitting layer 11, an electron injection layer 12 and cathodes 13, was formed on the upper surface of the functional film laminate.

The results of the evaluation of the foregoing four devices according to the first and second embodiments (E1 and E2) and comparative examples 1 and 2 (C1 and C2) are listed in Table 1. In table 1 and hereinafter, the functional film laminates according to the first and second embodiments and the comparative example 2 and the protection layer according to the comparative example 1 will be

referred to collectively as the "intermediate films". The evaluation methods for the respective terms and interpretations of the results will be described later.

Table 1

	Thickness of the intermediate films (μ m)	Flatness (μ m)	Angles of visibility	Life of the devices (hr.)	Influences to the flouresecent materials
E1	20	$<\pm 1$	$>\pm 60^{\circ}$	>10000	None
E2	20	$<\pm 1$	$>\pm 60^{\circ}$	>10000	None
C1	50	$>\pm 5$	$<\pm 30^{\circ}$	<100	Caused
C2	20	$<\pm 1$	$>\pm 60^{\circ}$	<100	None

The thickness of the functional film laminate is defined as the height "a" from the upper surface of the transparent substrate 7 to the upper surface of the hard coat layer 3 as described in Figure 2. As the results for the first and second embodiments clearly indicate, the fluorescent materials are protected by the very thin functional film laminate according to the invention.

Height deviation of the surface of the intermediate film on the colour conversion filter was examined with a surface roughness meter (DEKTAK II A supplied from ULVAC JAPAN Ltd.). As the results for the first and second embodiments clearly indicate, the height deviation of the surface of the bonded functional film laminate according to the invention is within $\pm 1 \mu$ m. While, the height deviation of the surface of the protection layer formed by resin coating is outside the range of $\pm 5 \mu$ m.

The angle of visibility is defined as the angle range, outside which light leakage to the adjacent fluorescent layers for the other colours is observed when the organic EL element emits monochromatic light. In the devices of the first and second embodiments and the comparative example 2 in which the functional film laminate is thin (20 μ m), the angles of visibility were more than 60° on both sides, which poses no problems on practical use of the devices. While, the angle ofvisibility of the comparative example 1, whose protection layer is thick (50 μ m), was less than 30° on both sides.

Figure 4 is a pair of curves plotting the maximum dark spot sizes in the devices according to the first embodiment and the comparative example 2 with elapse of time. The devices according to the first embodiment and the comparative example 2 were placed in a nitrogen gas stream and growth of dark spots in the respective light-emitting portions (2 mm) was observed under an optical microscope. It has been confirmed that the hard coat layer 3, arranged atop the functional film laminate according to the invention and working as a gas barrier, blocks the chemical species, which deteriorate the organic EL element, such as moisture and monomers originally contained in or yielded from the fluorescent materials and the coupling layer by the heat generated in driving the device and that the hard coat layer according to the invention effectively prevents the dark spots from growing.

The devices according to the embodiments and the comparative examples were placed in a nitrogen gas stream and the changes of the CIE colour coordinate when the devices emit monochromatic light were examined. It has been confirmed that the fluorescent materials protected by the functional film laminate according to the invention conduct stable operations after the completion of the organic EL device.

As explained above, the present invention facilitates providing a polychromatic display device which exhibits a wide angle of visibility and a long light-emitting life and manufacturing such a polychromatic display device easily with cheap manufacturing costs.

CLAIMS

1. A polychromatic organic electroluminescent device comprising:

a colour conversion filter comprising a transparent substrate and fluorescent layers on the transparent substrate, the fluorescent layers being spaced apart from each other;

an organic electroluminescent element comprising an organic light-emitting layer; and

a functional film laminate interposed between the colour conversion filter and the organic electroluminescent element;

the colour conversion filter and the organic electroluminescent element being arranged such that the fluorescent layers absorb the light from the organic light-emitting layer and emit light in their respective colours; the functional film laminate comprising a transparent base film, a transparent hard coat layer on the upper surface of the transparent base film and a coupling layer on the lower surface of the transparent base film;

the hard coat layer being transparent for the visible light, the hard coat layer exhibiting a gas blocking capability and hardness enough to endure the formation of the organic electrolumineacent element on the upper surface thereof;

the coupling layer comprising a photo-curing resin or a thermosetting resin which hardens at 150°C or lower.

2. The polychromatic organic electroluminescent device according to Claim 1, wherein one of the fluorescent layers is replaced by a colour filter for adjusting the colour and the colour purity of the light emitted from the organic light-emitting layer.

3. The polychromatic organic electroluminescent device according to Claim 1, wherein the functional film laminate further comprises transparent electrodes on the hard coat layer.

4. The polychromatic organic electroluminescent device according to Claim 1, wherein the hard coat layer comprises a resin layer or an inorganic oxide layer the resin layer or the inorganic oxide layer exhibiting pencil hardness H or more.

5. The photochomatic organic electroluminescent device according to Claim 1, wherein the coupling layer contains a resin which hardens by the visible light.

6. The polychromatic organic electrolumimescent device according to Claim 1, wherein the coupling layer contains a resin which thermally cross-links at 150°C or lower.

7. The polychromatic organic electroluminescent device according to Claim 1, wherein the base film is from 1 to 50 μm in thickness.

8. The polychromatic organic electroluminescent device according to Claim 1, wherein the fluorescent layers include a red fluorescent layer which converts the light from the organic electroluminescent element to red light, a green fluorescent layer which converts the light from the organic electroluminescent element to green light and a blue fluorescent layer which converts the light from the organic electroluminescent element to blue light.

9. A method of manufacturing a polychromatic organic electroluminescent device which includes a colour conversion filter including a transparent substrate and fluorescent layers formed on a transparent substrate and spaced apart from each other; an organic electroluminescent element including an organic light-emitting layer; and a functional film laminate interposed between the colour conversion filter and the organic electroluminescent element; the colour conversion filter and the organic electroluminescent element being arranged such that the fluorescent layers absorb the light from the organic light-emitting layer and emit light in their respectivecolours; the functional film laminate including a transparent base film, a transparent hard coat layer on the upper surface of the transparent base film and a coupling layer on the lower surface of the transparent base film; the hard coat layer being transparent for the visible light, the hard coat layer exhibiting a gas blocking capability and hardness enough to endure the formation of the organic electroluminescent element on its upper surface; the coupling layer comprising a photo-curing resin or a thermosetting resin which hardens at 150°C or

lower, the method comprising the steps of:

bonding the functional film laminate onto the upper surface of the colour conversion filter; and

arranging the organic electroluminescent element on the upper surface of the functional film laminate,

10. A polychromatic organic electroluminescent device substantially as describe herein with reference to and as shown in Figures 1 to 4 and Figures 7 to 10 of the accompanying drawings.

11. A method of manufacturing a polychromatic organic electroluminescent device substantially as describe herein with reference to and as shown in Figures 1 to 4 and Figures 7 to 10 of the accompanying drawings.

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Claims searched: All **Date of search:** 18 January 1999

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Databases searched:

UK Patent Office collections, including GB, EP, WO & US patent specifications, in:

UK Cl (Ed.Q): H1K(KEAL,KEAM)

Int Cl (Ed.6): H01L

Other: ON LINE,W.P.I.

Documents considered to be relevant:

Category	Identity of document and relevant passage	Relevant to claims
	NONE	

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